

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

Kiyoshige MURAOKA et al.

Group Art Unit: 1733

Serial Number: 10/679,406

Examiner: Geoffrey L. Knable

Filed: October 7, 2003

For: TUBELESS TIRE

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents  
P.O. Box 1450  
Alexandria, Virginia 22313-1450

Sir:

Kiyoshige Muraoka residing at 4-3-2, Oyashikidori, Nagata-ku, Kobe-shi, Hyogo, Japan duly deposes and says:

1. That he graduated from Department of Applied Chemistry, School of Engineering, University of Hyogo, Hyogo, Japan, in the year 1983, and he received the degree of Master of Process Engineering from Graduate School of Osaka University, Osaka, Japan in the year 1985;
2. That since 1985, he has been employed in the capacity of Sumitomo rubber Industries, Ltd.;
3. That from 1985 he has been engaged in development of rubber compositions for tires.;
4. That he has read and is familiar with the instant application for United States Letters Patent and Office Action thereto mailed April 20, 2006.; and

5. That he has made experiments in order to show that especially all of air permeability coefficient, rate of decrease in pressure and rate of decrease in tire pressure after machining are deteriorated when an inorganic layered compound is not contained in a gas barrier layer.

**<Process for preparing coating solution for gas barrier layer>**

Coating solution 1 and 2 are prepared in the same manner as described in the present specification.

**Coating solution 1**

Synthetic mica (tetrasilylic mica (Na-Ts); available from Topy Industries, Limited) was dispersed in ion exchanged water so as to become 0.65 % by weight to obtain an inorganic layered compound dispersion solution (solution A). The average particle size of the synthetic mica (Na-Ts) was 977 nm and the average aspect ratio was 1043. Polyvinyl alcohol (PVA210; available from Kuraray Co., Ltd., weight percentage of hydrogen bond groups or ionic groups per weight unit of resin: 38 %, hydrolyzation degree: 88.5 %, polymerization degree: 1000) was dissolved in ion exchanged water so as to become 0.325 % by weight to obtain a resin solution (solution B). Solution A and solution B were mixed so that the respective solid content ratio (volume ratio) was inorganic layered compound/resin = 3/7 to obtain coating solution 1 for the gas barrier layer.

**Coating solution 2**

The same polyvinyl alcohol (PVA210; available from Kuraray Co., Ltd.,) as that used in coating solution 1 for the gas barrier layer was dissolved in ion exchanged water so as to become

0.325 % by weight to obtain coating solution 2 for the gas barrier layer.

**<Preparation of rubber composition>**

Rubber compositions are prepared in the same manner as described in the present specification.

Each sample rubber composition (rubber composition C and U) was prepared with the materials and processing methods described below.

**(Materials)**

NR: RSS #3 available from Tech Bee Hang Co., Ltd.,

Br-IIR: Exxon Bromobutyl 2255

Sodium-bentonite: Kunipia F (clay-type mineral with a primary particle size of 100 to 2000 nm, average aspect ratio of 320 and swellability of at least 45 ml/2 g) available from Kunimine Industries

GPF: Seast V available from Tokai Carbon Co., Ltd.,

HAF: Diablock H available from Mitsubishi Chemical Corporation

Silica: Ultrasil VN3 (N<sub>2</sub>SA: 210 m<sup>2</sup>/g) available from Degussa Co.

Silane coupling agent: Si69 (bis(3-triethoxysilylpropyl)tetrasulfide) available from Degussa Co.

Resin: ESCOREZ 1102 available from Esso

Oil A: JOMO Process X-140 available from Japan Energy Corporation

Stearic Acid: Stearic acid available from NOF Corporation

Zinc oxide: Zinc Oxide Type 1 available from Mitsui Mining and Smelting Co., Ltd.,

Sulfur: powdery sulfur available from Tsurumi Chemicals Co., Ltd.,

Vulcanization Accelerator NS: Nocceler NS

(N-tert-butyl-2-benzothiazylsufenamide), available from Ouchi Shinko Chemical Industrial Co., Ltd.,

Vulcanization Accelerator D: Nocceler D (N,N'-diphenyl guanidine), available from Ouchi Shinko Chemical Industrial Co., Ltd.,  
(Processing method)

According to the composition formula shown in Table 1, the materials other than sulfur, zinc oxide and the vulcanization accelerator were kneaded in a BR-type Banbury to prepare a master batch and then the master batch and sulfur, zinc oxide and the vulcanization accelerator were kneaded with an 8 inch roll to obtain each sample rubber composition (rubber composition C and U). The obtained rubber composition was press vulcanized at 170°C for 15 minutes to obtain a vulcanized object.

TABLE 1

Rubber Composition	C	U	Inner Liner Composition
Composition (parts by weight)			
NR	40	40	50
Br-IIR	60	60	50
GPF	50	—	50
HAF	—	20	—
Sodium-bentnrite	—	5	—
Resin	2	2	2
Oil A	10	10	15
Silica	—	20	—
Silane Coupling Agent	—	2	—
Stearic Acid	2	2	2
Zinc Oxide	3	3	3
Sulfur	1.5	1.5	1.5
Vulcanization Accelerator NS	1	1	1
Vulcanization Accelerator D	—	1	—

Example 1 and Comparative Example 6

<Preparation of rubber composition>

A polyurethane anchor coating (Adocoat AD 335A/CAT10L = 15/1 (weight ratio); available from Toyo Morton Co., Ltd.) was applied on the various vulcanized rubber composition (rubber composition C) shown in TABLE 2. Then, after the various coating solutions for the gas barrier layer were formed into a film by casting, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 1  $\mu$ m

for the anchor coat layer and approximately 2  $\mu\text{m}$  for the gas barrier layer. The following air permeability evaluation was conducted with respect to the rubber. The results are shown in TABLE 2.

**<Preparation of tire>**

A 195/65R14 tire was prepared using the above rubber composition as the inner liner layer and then vulcanized.

The anchor coating and then the coating solution for the gas barrier layer were applied to the inner face of the inner liner layer of the vulcanized tire using a certain spray gun and then dried. Then, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 1  $\mu\text{m}$  for the anchor coat layer and approximately 2  $\mu\text{m}$  for the gas barrier layer. The following evaluation of the rate of decrease in tire pressure was conducted with respect to the tire. The results are shown in TABLE 2.

**Example 16 and Comparative Example 39**

**<Preparation of rubber composition>**

After coating solutions 1 or 2 were formed into a film by casting on the various rubber samples obtained by rubber composition U, according to the compounding formula shown in TABLE 2, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 5  $\mu\text{m}$ . The air permeability evaluation was conducted with respect to the rubber. The results are shown in TABLE 2.

In each Examples and Comparative Examples, "1" indicates that coating solution 1 for the gas barrier layer was formed into a film and "2" indicates that coating solution 2 for the gas barrier

layer was formed into a film, in the gas barrier composition of TABLE 2.

<Preparation of tire>

A 195/65R14 tire was prepared using the above rubber composition as the carcass layer and then vulcanized. In Example 16, an inner liner layer was not formed. The coating solution was applied to the inner face of the vulcanized tire using a certain spray gun and then dried. Then, thermal treatment was conducted for 10 minutes at 100°C. The film thickness obtained by the treatment was approximately 2  $\mu\text{m}$ . The following evaluation of the rate of decrease in tire pressure was conducted with respect to the tire. The results are shown in TABLE 2.

<Evaluation as rubber composition>

Rubber compositions are evaluated in the same manner as described in the present specification.

(Air permeability evaluation

The air permeability coefficient was measured according to JIS K7126 "Testing method of gas permeability of plastic films and sheets (method A)" with air as the test gas (nitrogen:oxygen = 8:2) and 25°C as the test temperature. The air permeability is decreased the lower the number value is.

<Evaluation as tire>

Tires are evaluated in the same manner as described in the present specification.

(Evaluation of rate of decrease in tire pressure)

The above tire with an initial pressure of 100 kPa was left

for 3 months in room temperature of 25°C under a no-load condition and the pressure was measured every 4 days. When  $t$  is the number of days passed,  $P_0$  is the initial pressure and  $P_t$  is the pressure measured when  $t$  days have passed, the value of  $\alpha$  is found by using function:

$$P_t/P_0 = \exp(-\alpha t).$$

The obtained  $\alpha$  and  $t = 30$  were substituted into the following equation and  $\beta$  was obtained. The value of  $\beta$  was assumed to be the rate of decrease in pressure per month (%/month).

$$\beta = \{1 - \exp(-\alpha t)\} \times 100$$

The rate of decrease in pressure of the tire is preferably at most 2.5 (%/month). When more than 2.5 %, decrease in tire pressure becomes noticeable.

(Tire durability test)

An indoor drum test was conducted using a specific tire under conditions of running speed of 80 km/hour, pressure of 190 kPa and load of 464 kg. The running results are represented by the distance run from when running is begun until abnormality such as burst or change in the tire appearance (swelling) is observed. The durability is superior the longer the running distance is.

(Pressure test and evaluation of appearance after testing with tire machine)

An indoor drum test was conducted using a specific tire under conditions of running speed of 80 km/hour, pressure of 190 kPa and load of 646 kg and the tire was run for 30,000 km. The appearance of the inner liner layer was checked for tears and wrinkles in the gas barrier layer thin film. Then, with respect to the tire, the

test for the rate of decrease in tire pressure was conducted again.

TABLE 2

	Ex. 1	Com. Ex. 6	Ex. 16	Com. Ex. 39
Rubber Composition	C	C	U	U
Gas Barrier Layer Composition	1	2	1	2
Air Permeability Coefficient ( $\times 10^{-11}$ cc·cm/cm <sup>2</sup> ·sec·cmHg)	7	11	4	6
Inner Liner (Rubber Composition Z) (Air Permeability Coefficient: $19 \times 10^{-11}$ cc·cm/cm <sup>2</sup> ·sec·cmHg)	Present	Present	None	None
Rate of Decrease in Pressure (%/month)	2.1	2.3	2.0	2.1
Tire Weight	9.13	9.14	8.61	8.60
Results of Durability Test (10,000 km)	3.0 No	3.0 No	3.0 No	3.0 No
	Breakage	Breakage	Breakage	Breakage
Rate of Decrease in Tire Pressure after Machining	2.1	2.5	2.1	2.3
Appearance of Gas Barrier layer Inside Tire	-	-	No Wrinkles	Few Wrinkles

#### Results and Discussions

As evident from the results of Table 2, especially all of air permeability coefficient, rate of decrease in pressure and rate of decrease in tire pressure after machining are deteriorated when an inorganic layered compound is not contained in a gas barrier layer. On the other hand, all of air permeability coefficient, rate of decrease in pressure and rate of decrease in tire pressure after machining are improved when an inorganic layered compound is contained in a gas barrier layer.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 29th day of August, 2007

by Kiyoshige Muraoka  
Kiyoshige Muraoka

We, the undersigned witnesses, hereby acknowledge that Kiyoshige Muraoka is personally known to us and did execute the foregoing Declaration in our presence on:

Date: August 29, 2007

Witness

Yutaka Sakon

Date: August 29, 2007

Witness

Naoki Fujita